



(12) **United States Patent**  
**Xue et al.**

(10) **Patent No.:** **US 9,425,375 B2**  
(45) **Date of Patent:** **Aug. 23, 2016**

(54) **METHOD FOR MAKING  
HIGH-TEMPERATURE SUPERCONDUCTING  
FILM**

(71) Applicants: **Tsinghua University**, Beijing (CN);  
**Institute of Physics, Chinese Academy  
of Sciences**, Beijing (CN)

(72) Inventors: **Qi-Kun Xue**, Beijing (CN); **Xu-Cun  
Ma**, Beijing (CN); **Li-Li Wang**, Beijing  
(CN); **Xi Chen**, Beijing (CN); **Jin-Feng  
Jia**, Beijing (CN); **Ke He**, Beijing (CN);  
**Shuai-Hua Ji**, Beijing (CN); **Wen-Hao  
Zhang**, Beijing (CN); **Qing-Yan Wang**,  
Beijing (CN); **Zhi Li**, Beijing (CN)

(73) Assignees: **Tsinghua University**, Beijing (CN);  
**Institute of Physics, Chinese Academy  
of Sciences**, Beijing (CN)

(\*) Notice: Subject to any disclaimer, the term of this  
patent is extended or adjusted under 35  
U.S.C. 154(b) by 393 days.

(21) Appl. No.: **14/316,823**

(22) Filed: **Jun. 27, 2014**

(65) **Prior Publication Data**  
US 2015/0380130 A1 Dec. 31, 2015

(51) **Int. Cl.**  
**H01L 39/24** (2006.01)  
**H01L 39/12** (2006.01)  
**H01B 13/00** (2006.01)  
**H01B 12/06** (2006.01)

(52) **U.S. Cl.**  
CPC ..... **H01L 39/2403** (2013.01); **H01L 39/12**  
(2013.01); **H01B 12/06** (2013.01); **H01B**  
**13/0036** (2013.01)

(58) **Field of Classification Search**  
CPC ..... H01B 12/06; H01B 13/0036  
USPC ..... 505/410  
See application file for complete search history.

(56) **References Cited**

U.S. PATENT DOCUMENTS

2013/0196856 A1\* 8/2013 Li ..... H01L 39/125  
505/191

OTHER PUBLICATIONS

Qingyan Wang et al.; "Interface-induced high-temperature supercon-  
ductivity in single unit-cell FeSe films on SrTiO<sub>3</sub>"; Chinese Physics  
Letters, vol. 29, No. 3, 2012-03.

\* cited by examiner

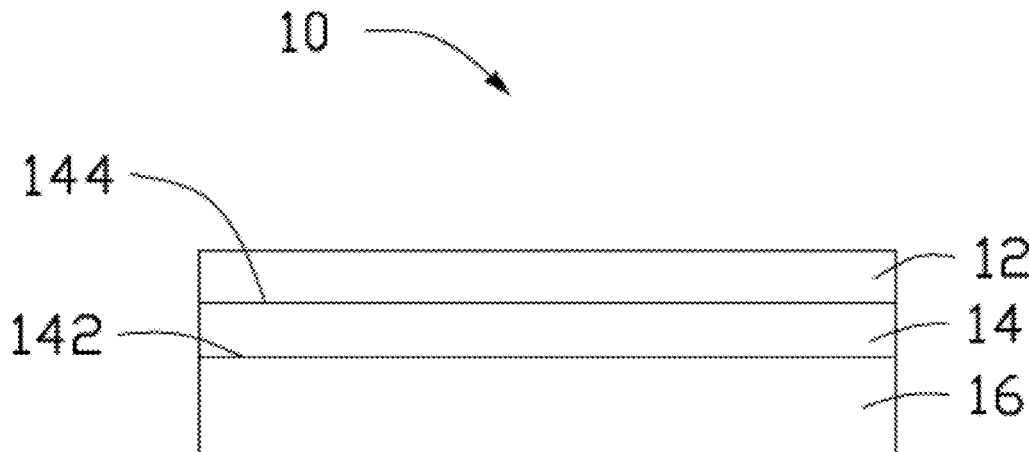
*Primary Examiner* — Paul Wartalowicz

(74) *Attorney, Agent, or Firm* — ScienBiziP, P.C.

(57) **ABSTRACT**

A method for making a high-temperature superconducting  
film includes loading a SrTiO<sub>3</sub> substrate in an ultra-high  
vacuum system. A single crystalline FeSe layer is grown on a  
surface of the SrTiO<sub>3</sub> substrate by molecular beam epitaxy. A  
protective layer with a layered crystal structure is grown by  
molecular beam epitaxy and covering the single crystalline  
FeSe layer.

**20 Claims, 7 Drawing Sheets**



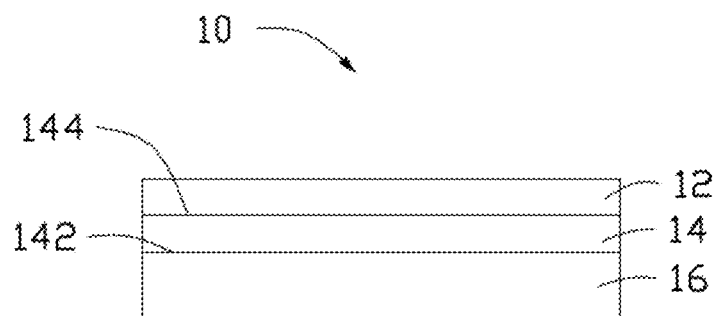


FIG. 1

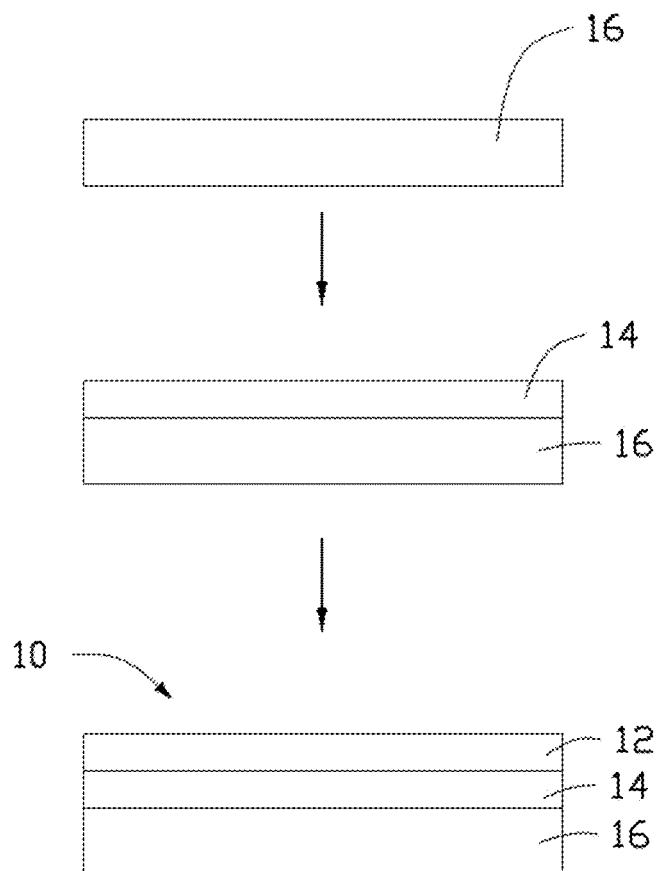


FIG. 2

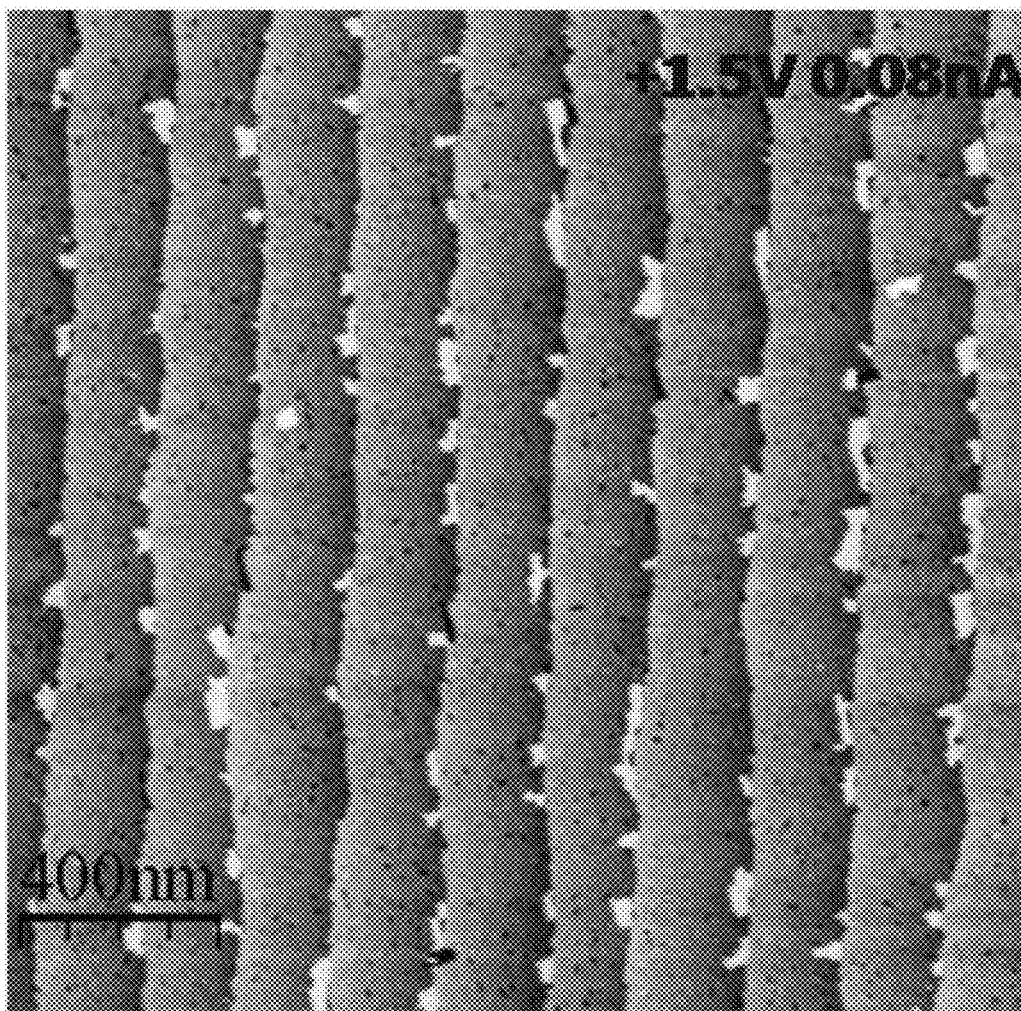


FIG. 3

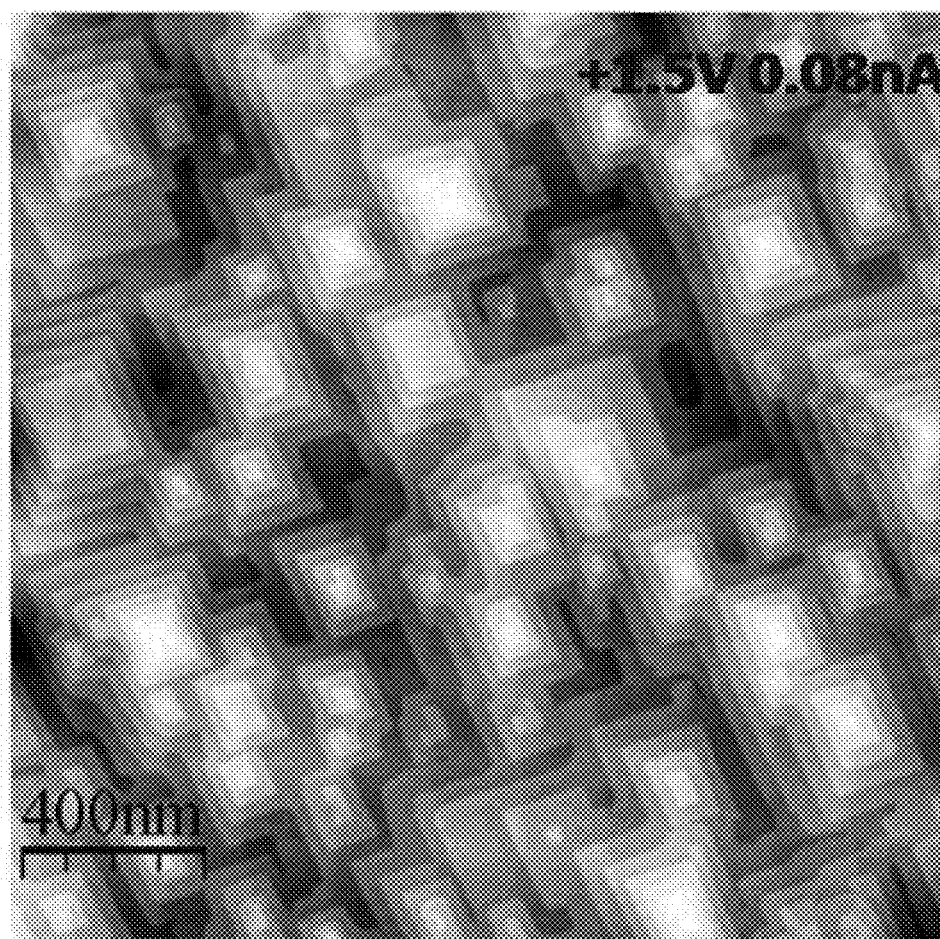


FIG. 4

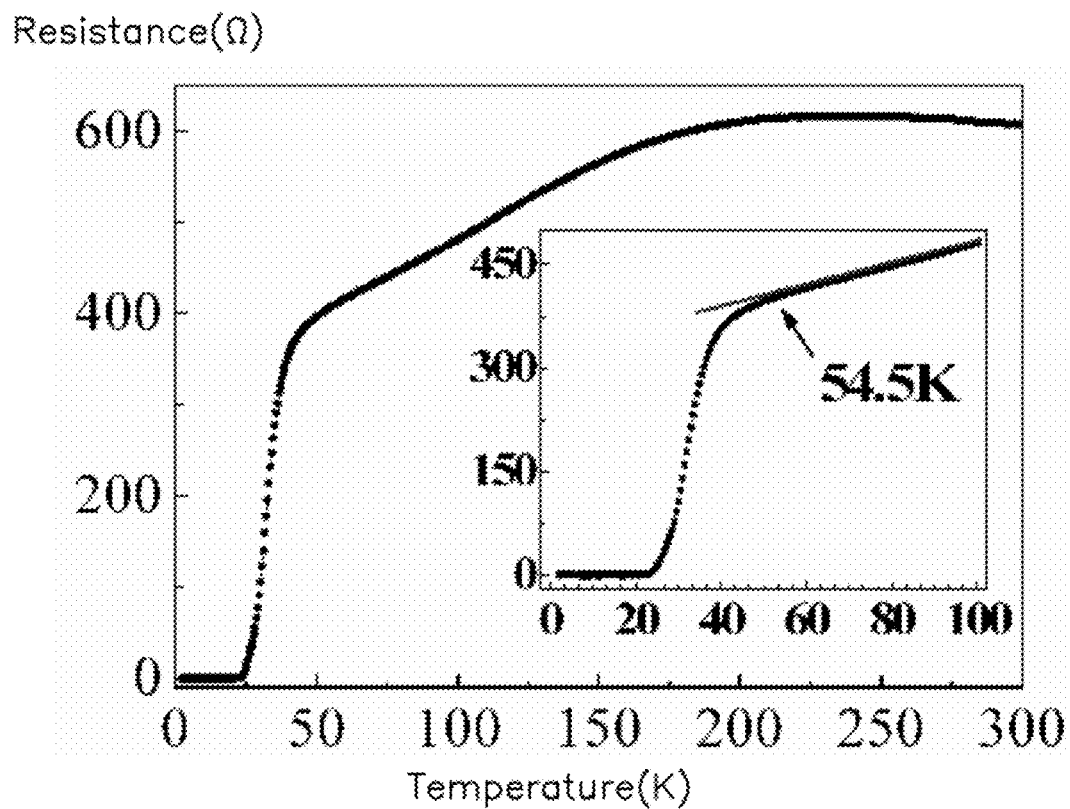


FIG. 5

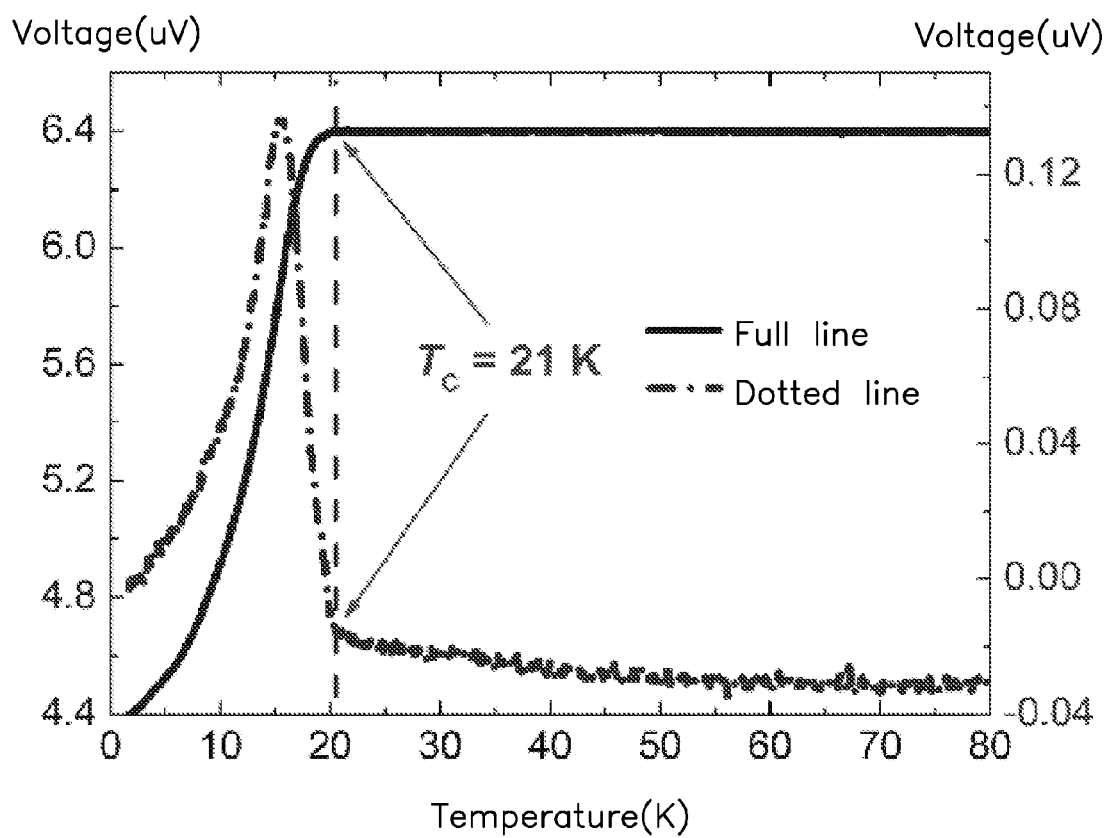


FIG. 6

Critical current  
density(A/cm<sup>2</sup>)

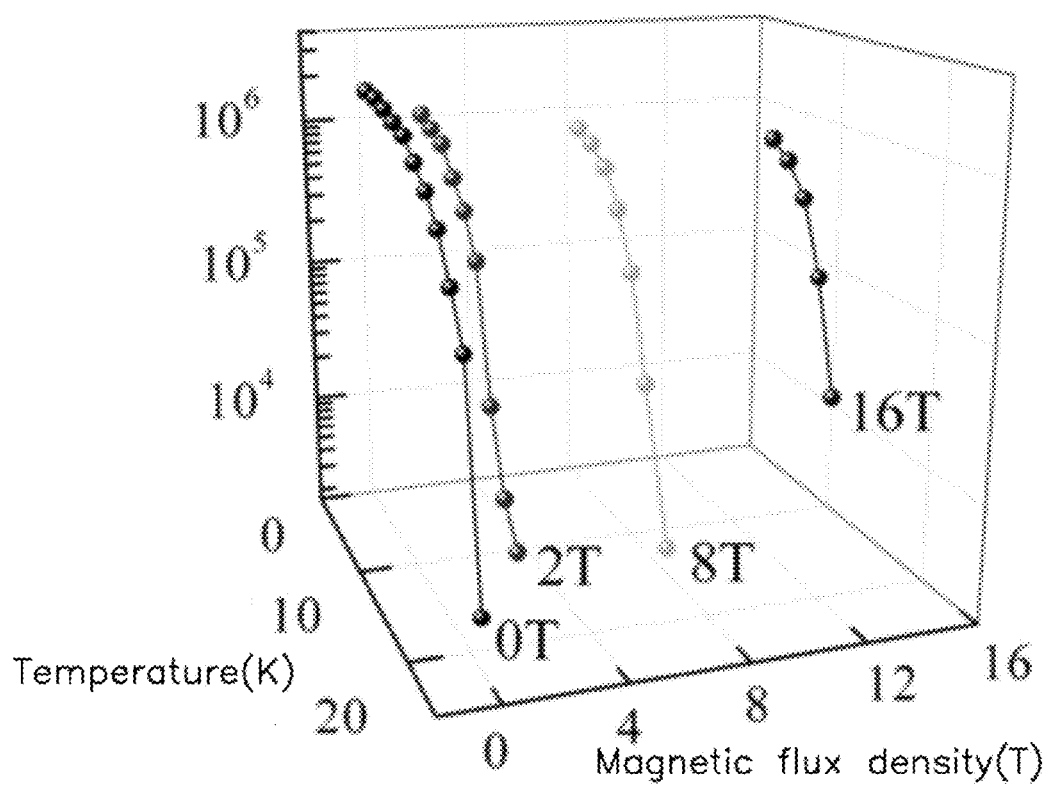


FIG. 7



1

## METHOD FOR MAKING HIGH-TEMPERATURE SUPERCONDUCTING FILM

### CROSS-REFERENCE TO RELATED APPLICATIONS

This application is related to commonly-assigned application entitled, "HIGH-TEMPERATURE SUPERCONDUCTING FILM", concurrently filed Ser. No. 14/316,822. Disclosures of the above-identified applications are incorporated herein by reference.

### FIELD

The present application relates to a method for making high-temperature superconducting film.

### BACKGROUND

High temperature superconductor is an unconventional superconductor which cannot be explained by conventional BCS theory. In 1986, Muler and Bednorz firstly discovered cuprate high-temperature superconductors. In 2008, physicists in Japan firstly discovered iron-based high-temperature superconductors. This provides a new system for studying mechanism of superconductivity and gives inspiration to explore other high-temperature superconducting materials.

The iron-based high-temperature superconductors and cuprate high-temperature superconductors have some resemblances in structure. All of them own layered heterostructures with one conducting layer sandwiched between two insulating layers.

In addition, high temperature superconducting materials usually are synthesized by high temperature reaction, single crystalline or polycrystalline. However, bulk materials are known to suffer from great fluctuations in stoichiometry, disorder, and clustering pathologies with many defects and impurities. At present, iron-based high-temperature superconducting films can be prepared by Pulsed Laser Deposition (PLD). However, these iron-based high-temperature superconducting films usually comprise inhomogeneous structure and many impurities, which may affect studying the mechanism of high-temperature superconductivity.

What is needed, therefore, is to provide a method for making high-temperature superconducting film which can overcome the shortcomings as described above.

### BRIEF DESCRIPTION OF THE DRAWINGS

Implementations of the present technology will now be described, by way of example only, with reference to the attached figures, wherein:

FIG. 1 is a schematic view of one embodiment of a high-temperature superconducting film.

FIG. 2 is a schematic process flow of one embodiment of a method for making the high-temperature superconducting film of FIG. 1.

FIG. 3 shows a scanning tunneling microscope (STM) topographic image of a single crystalline FeSe layer of the high-temperature superconducting film of FIG. 1.

FIG. 4 shows a scanning tunneling microscope (STM) topographic image of the single crystalline FeSe layer of FIG. 3 covered by a protective layer.

FIG. 5 shows an ex situ electrical transport measurement of the high-temperature superconducting film of FIG. 1.

2

FIG. 6 shows a diamagnetism measurement of the high-temperature superconducting film of FIG. 1.

FIG. 7 shows a critical current density measurement of the high-temperature superconducting film of FIG. 1.

### DETAILED DESCRIPTION

It will be appreciated that for simplicity and clarity of illustration, where appropriate, reference numerals have been repeated among the different figures to indicate corresponding or analogous elements. In addition, numerous specific details are set forth in order to provide a thorough understanding of the embodiments described herein. However, it will be understood by those of ordinary skill in the art that the embodiments described herein can be practiced without these specific details. In other instances, methods, procedures and components have not been described in detail so as not to obscure the related relevant feature being described. The drawings are not necessarily to scale and the proportions of certain parts may be exaggerated to better illustrate details and features. The description is not to be considered as limiting the scope of the embodiments described herein.

Several definitions that apply throughout this disclosure will now be presented.

The term "coupled" is defined as connected, whether directly or indirectly through intervening components, and is not necessarily limited to physical connections. The connection can be such that the objects are permanently connected or releasably connected. The term "outside" refers to a region that is beyond the outermost confines of a physical object. The term "inside" indicates that at least a portion of a region is partially contained within a boundary formed by the object. The term "substantially" is defined to be essentially conforming to the particular dimension, shape or other word that substantially modifies, such that the component need not be exact. For example, substantially cylindrical means that the object resembles a cylinder, but can have one or more deviations from a true cylinder. The term "comprising" means "including, but not necessarily limited to"; it specifically indicates open-ended inclusion or membership in a so-described combination, group, series and the like.

Referring to FIG. 1, an iron-based high-temperature superconducting film 10 includes a SrTiO<sub>3</sub> substrate 16, a single crystalline FeSe layer 14, and a protective layer 12. The single crystalline FeSe layer 14 and the protective layer 12 are epitaxially grown on the SrTiO<sub>3</sub> substrate 16 via a typical layer-by-layer mode. The single crystalline FeSe layer 14 is sandwiched between the SrTiO<sub>3</sub> substrate 16 and the protective layer 12. A first interface 142 between the SrTiO<sub>3</sub> substrate 16 and the single crystalline FeSe layer 14 is atomically smooth. A second interface 144 between the single crystalline FeSe layer 14 and a protective layer 12 is also atomically smooth, basically no ion intermixing on atomic scale.

The SrTiO<sub>3</sub> substrate 16 has a relatively high dielectric constant at low temperature, which is beneficial to screen interaction between carriers. The SrTiO<sub>3</sub> substrate 16 can be insulating, which is good for measuring the superconducting transition temperature of the high-temperature superconducting film 10 by electrical transport measurement. A thickness of the SrTiO<sub>3</sub> substrate 16 can be in a range from about 0.2 millimeters (mm) to about 1.0 mm. In one embodiment, the thickness of the SrTiO<sub>3</sub> substrate 16 is about 0.5 mm.

The SrTiO<sub>3</sub> substrate 16 has a layered crystal structure, which is formed by alternatively stacking a titanium dioxide (TiO<sub>2</sub>) layer and a strontium oxide (SrO) layer. A crystal lattice of the SrTiO<sub>3</sub> substrate 16 in the (100) crystal plane is a tetragonal lattice, with a lattice constant of 0.3905 nm. The

lattice mismatch between the  $\text{SrTiO}_3$  substrate **16** and the single crystalline FeSe layer **14** is about 3%, which is good for high-quality single crystalline FeSe layer **14** grown on the (100) crystal plane of the  $\text{SrTiO}_3$  substrate **16**.

The single crystalline FeSe layer **14** has a layered crystal structure in a tetragonal lattice, with the lattice constant 0.378 nm in the (100) crystal plane. A thickness of the single crystalline FeSe layer **14** is in a range from about 1 unit-cell (UC) to about 5 UC. Along the (001) crystal direction, one UC thick FeSe film consists of a Se—Fe—Se triple layer, where two adjacent Fe atoms are bonded by covalent bonds in the Fe atom layer and each two Se atoms are bonded to the Fe atoms by covalent bonds above and below the planar Fe layer, respectively. Two adjacent triple layers are bonded by Van der Waals force. In one embodiment, the single crystalline FeSe layer **14** is a 1 UC thick FeSe single crystalline film.

The superconducting transition temperature is enhanced by interface effect at the first interface **142**. When the single crystalline FeSe layer **14** is 1 UC thick, the superconducting transition temperature reaches the highest value. When the thickness of the single crystalline FeSe layer **14** becomes thicker, the superconducting transition temperature of the high-temperature superconducting film **10** decreases because of proximity effect.

The protective layer **12** has a layered crystal structure with tetragonal lattice. The tetragonal lattice layered compound is composed by A and B, wherein A is a transition metal, B is selenium (Se), tellurium (Te), or sulfur (S). In one embodiment, the protective layer **12** is a single crystalline FeTe layer. The single crystalline FeTe layer is non-superconducting. A thickness of the single crystalline FeTe layer can be in a range from about 2 UC to about 10 UC. In one embodiment, the thickness of the single crystalline FeTe layer is 10 UC, to protect the single crystalline FeSe layer **14**. Thus, the single crystalline FeSe layer **14** is prevented from oxidation and adsorbing impurities such as water vapor in air.

For the protective layer **12** and the single crystalline FeSe layer **14**, their crystal structure is the same, and their lattice mismatch is less than 5%, consequently the second interface **144** formed between the protective layer **12** and the single crystalline FeSe layer **14** is atomically smooth. The protective layer **12** can fully cover the single crystalline FeSe layer **14**, and directly contact the whole single crystalline FeSe layer **14**.

The first interface **142** is in favor of the heterointerface of the FeSe/ $\text{SrTiO}_3$  to produce a strong interface-enhanced superconductivity effect. The second interface **144** can minimize external factors influencing the interface-enhanced superconductivity effect.

Referring to FIG. 2, one embodiment of a method for making the high-temperature superconducting film **10** of includes:

(S1) loading a  $\text{SrTiO}_3$  substrate **16** into an ultra-high vacuum (UHV) system;

(S2) growing a single crystalline FeSe layer **14** on a surface of the  $\text{SrTiO}_3$  substrate **16** by molecular beam epitaxy (MBE); and

(S3) growing a protective layer **14** with a layered crystal structure by MBE and covering the single crystalline FeSe layer **14**.

In the step (S1), before loading the  $\text{SrTiO}_3$  substrate **16** in an UHV system, the  $\text{SrTiO}_3$  substrate **16** is successively treated by deionized water, acid, and high temperature annealing in oxygen atmosphere, to obtain a single  $\text{TiO}_2$  terminated surface and atomically flat steps on the  $\text{SrTiO}_3$  substrate **16**. Thus, high-quality single crystalline FeSe layer **14** and an atomically smooth interface can be formed.

The  $\text{SrTiO}_3$  substrate **16** is treated by deionized water. The  $\text{SrTiO}_3$  substrate **16** can be kept in deionized water for about 40 min to about 80 min, wherein a temperature of the deionized water can be in a range from about 70 degrees Celsius to about 100 degrees Celsius. In one embodiment, the  $\text{SrTiO}_3$  substrate **16** is kept in the deionized water at about 80 degrees Celsius for about 60 min. The purpose of treating by deionized water is to promote the adsorption of more  $\text{OH}^-$  on a surface of the  $\text{SrTiO}_3$  substrate **16** and formation a strontium hydroxide ( $\text{Sr}(\text{OH})_2$ ) with SrO on the surface of the  $\text{SrTiO}_3$  substrate **16**.

The  $\text{SrTiO}_3$  substrate **16** is treated by acid. That is, the surface of the  $\text{SrTiO}_3$  substrate **16** can be etched by hydrochloric acid (HCl) or hydrofluoric acid (HF), to neutralize  $\text{Sr}(\text{OH})_2$  on the surface of the  $\text{SrTiO}_3$  substrate **16**. In one embodiment, the surface of the  $\text{SrTiO}_3$  substrate **16** is etched by hydrochloric acid. The  $\text{SrTiO}_3$  substrate **16** treated by deionized water is kept in hydrochloric acid for about 30 min to about 60 min, wherein a concentration of the hydrochloric acid can be in a range from about 8% to about 12%. In one embodiment, the  $\text{SrTiO}_3$  substrate **16** treated by deionized water is etched in hydrochloric acid for about 45 min, wherein the concentration of the hydrochloric acid is 10%.

The  $\text{SrTiO}_3$  substrate **16** is annealed at a high temperature in oxygen atmosphere. That is, after being etched by acid and dried by nitrogen, the  $\text{SrTiO}_3$  substrate **16** can be annealed in a pure oxygen atmosphere for about 2.5 hours to about 3.5 hours. The annealing temperature can be in a range from about 950 degrees Celsius to about 1000 degrees Celsius. In one embodiment, the  $\text{SrTiO}_3$  substrate **16** can be placed in a high temperature tube furnace. The  $\text{SrTiO}_3$  substrate **16** is annealed to high temperature in oxygen atmosphere, and cooled to room temperature. In one embodiment, the  $\text{SrTiO}_3$  substrate **16** is heated at 980 degrees Celsius for about 3 hours. Wherein a heating rate is about 0.833 degrees Celsius per minute (/min) at a temperature from about 50 degrees Celsius to about 105 degrees Celsius, the heating rate is about 9.210 degrees Celsius per minute at temperatures from about 105 degrees Celsius to about 980 degrees Celsius, a cooling rate is about 5 degrees Celsius per minute, and oxygen flow is about 38 milliliters per minute. The purpose of treating the  $\text{SrTiO}_3$  substrate **16** at high temperature with oxygen is to form regular steps on the surface of the  $\text{SrTiO}_3$  substrate **16** and keep the  $\text{SrTiO}_3$  substrate **16** insulating, which is propitious to prepare atomically flats on the FeSe film and performing the transport measurements.

The UHV facility is a system with base pressure less than  $10^{-7}$  Pa. In one embodiment, the base pressure is  $2.0 \times 10^{-8}$  Pa. A molecular beam epitaxy device is located in the UHV system.

After loading the  $\text{SrTiO}_3$  substrate **16** into the UHV system, the  $\text{SrTiO}_3$  substrate **16** is degassed at a temperature from about 600 degrees Celsius to about 650 degrees Celsius for a time in the range of from about 2.5 hours to about 3.5 hours, to remove the impurities absorbed on the surface of the  $\text{SrTiO}_3$  substrate **16** in an atmosphere. In one embodiment, the  $\text{SrTiO}_3$  substrate **16** is degassed at about 600 degrees Celsius for about 3 hours.

In the step (S2), in the process of growth by MBE, the  $\text{SrTiO}_3$  substrate **16** is held at a temperature from about 380 degrees Celsius to about 420 degrees Celsius. A Fe source and a Se source are provided, wherein a flux ratio of the Fe source and the Se source can be in a range from about 1:10 to about 1:20. The evaporated temperature of the Fe source can be in a range from about 1000 degrees Celsius to about 1100 degrees Celsius. The evaporated temperature of the Se source can be in a range from about 130 degrees Celsius to about 150

degrees Celsius. The growth rate of the single crystalline FeSe layer **14** is about 0.25 UC per minute. In one embodiment, the growth rate ratio of the Fe source and the Se source is about 1:10, the SrTiO<sub>3</sub> substrate **16** is held at about 400 degrees Celsius, the evaporated temperature of the Fe source is about 1080 degrees Celsius, and the evaporated temperature of the Se source is about 138 degrees Celsius. The parameters above can improve the quality of the single crystalline FeSe layer **14**, further increasing the superconducting transition temperature of the high-temperature superconducting film **10**.

When the growth rate of the single crystalline FeSe layer **14** is about 0.25 UC per minute, a thickness of the single crystalline FeSe layer **14** can be controlled by the growth time. When the thickness is about 1 UC, the growth time is about 4 min. When the thickness is about 5 UC, the growth time is about 20 min.

After growing the single crystalline FeSe layer **14**, an entirety including the SrTiO<sub>3</sub> substrate **16** and the single crystalline FeSe layer **14** is annealed at a temperature from about 450 degrees Celsius to about 550 degrees Celsius for from about 20 hours to about 30 hours. In one embodiment, the entirety including the SrTiO<sub>3</sub> substrate **16** and the single crystalline FeSe layer **14** is annealed for about 25 hours at about 500 degrees Celsius. The purpose of annealing the entirety including the SrTiO<sub>3</sub> substrate **16** and the single crystalline FeSe layer **14** is to remove redundant Se and improving a strong bond between the single crystalline FeSe layer **14** and the SrTiO<sub>3</sub> substrate **16**, which is good for the high-temperature superconducting film **10** to obtain a stronger interface enhancement superconducting effect.

In the step (S3), the protective layer **12** is a single crystalline FeTe layer. In the process of growth by MBE, the SrTiO<sub>3</sub> substrate **16** is held at a temperature from about 310 degrees Celsius to about 330 degrees Celsius. A Fe source and a Te source are provided, wherein a flux ratio of the Fe source and the Te source can be in a range from about 1:10 to about 1:20. An evaporated temperature of the Fe source can be in a range from about 1000 degrees Celsius to about 1100 degrees Celsius. An evaporated temperature of the Te source can be in a range from about 240 degrees Celsius to about 280 degrees Celsius. A growth rate of the single crystalline FeTe layer is about 0.25 UC per minute. In one embodiment, the flux ratio of the Fe source and the Te source is about 1:10, the SrTiO<sub>3</sub> substrate **16** is held at about 320 degrees Celsius, the evaporated temperature of the Fe source is about 1070 degrees Celsius, and the evaporated temperature of the Te source is about 248 degrees Celsius. Under above parameters, high quality single crystalline FeTe layer can be prepared. The single crystalline FeSe layer **14** can be protected by the single crystalline FeTe layer.

When the growth rate of the single crystalline FeTe layer is about 0.25 UC per minute, a thickness of the single crystalline FeTe layer can be controlled by the growing time. When the thickness is about 2 UC, the growing time is about 8 min. When the thickness is about 10 UC, the growth time is about 40 min.

Referring to FIG. 3, an STM topographic image of the single crystalline FeSe layer **14** without the protective player **12** is shown, wherein the thickness of the single crystalline FeSe layer **14** is 1 UC. The single crystalline FeSe layer **14** has regular steps, originating from the SrTiO<sub>3</sub> substrate **16**, as shown in FIG. 3.

Referring to FIG. 4, an STM topographic image of the single crystalline FeSe layer **14** covered by the protective layer **12** is shown, wherein the thickness of the single crys-

talline FeTe layer is 10 UC. The single crystalline FeSe layer **14** is uniformly and completely covered by the single crystalline FeTe layer.

Referring to FIG. 5, an ex situ electrical transport measurement of the high-temperature superconducting film **10** is shown, wherein the thickness of the single crystalline FeSe layer **14** is 1 UC, and the thickness of the single crystalline FeTe layer is 10 UC. Since the SrTiO<sub>3</sub> substrate **16** is insulating, and neither the protective FeTe layer **12** nor the STO substrate **16** becomes superconducting due to proximity effect, so the superconductivity is confined to the 1 UC single crystalline FeSe layer **14**. As shown in FIG. 5, the resistance of high-temperature superconducting film **10** drops with decreasing temperature, and exhibits superconducting transition at low temperature. An onset temperature of the superconducting transition can be about 54.5 K. The resistance of the high-temperature superconducting film **10** completely reduces to zero at about 24 K.

Referring to FIG. 6, a diamagnetism measurement of the high-temperature superconducting film **10** is shown, wherein the thickness of the single crystalline FeSe layer **14** is 1 UC, and the thickness of the single crystalline FeTe layer is 10 UC. The real part of the inductance voltage is represented by a solid line, and the imaginary part of the inductance voltage is represented by a dotted line. The high-temperature superconducting film **10** appears to show perfect diamagnetism at a temperature about 21 K, which corresponds to the temperature when the resistance becomes zero as shown in FIG. 5.

Referring to FIG. 7, it shows a critical current density measurement of the high-temperature superconducting film **10**, wherein the thickness of the single crystalline FeSe layer **14** is 1 UC, and the thickness of the single crystalline FeTe layer is 10 UC. In zero field, the critical current density of the high-temperature superconducting film **10** is about 10<sup>6</sup> ampere per square centimeter (A/cm<sup>2</sup>) at 12 K. The critical current density of the high-temperature superconducting film **10** still remains about 10<sup>5</sup> A/cm<sup>2</sup> at 8 K and 16 T.

In summary, the lattice mismatch between SrTiO<sub>3</sub> and the single crystalline FeSe is small, wherein the SrTiO<sub>3</sub> (100) functions as a substrate. Thus, FeSe can be grown by MBE on the surface of SrTiO<sub>3</sub> in a layer-by-layer mode. The SrTiO<sub>3</sub> substrate **16** has a high dielectric constant at low temperature, which is beneficial to screen interaction between carriers and get strong FeSe/SrTiO<sub>3</sub> interface-enhanced superconducting effect. Furthermore, the single crystalline FeSe layer **14** can be atomically flat and prevented from oxidation and adsorbing impurities in air by single crystalline FeTe layer **12** as protective layer, which results in reducing the electron scattering at the interface between the single crystalline FeSe layer **14** and the protective layer **12**, preserving the superconductivity of the single crystalline FeSe layer **14**. Both the interface between the single crystalline FeSe layer **14** and the protective layer **12** and the interface between the single crystalline FeSe layer **14** and the SrTiO<sub>3</sub> substrate **16** are atomically smooth. Moreover, the onset temperature of the superconducting transition of the high-temperature superconducting film **10** is no less than 54 K. The critical current density of the high-temperature superconducting film **10** is about 10<sup>6</sup> A/cm<sup>2</sup> at 12 K. In addition, 1-UC FeSe on SrTiO<sub>3</sub> exhibits different electronic structures and thus different superconductivity mechanisms from those of bulk FeSe, which renders additional support to the interface enhanced electron-phonon coupling effect. The FeSe/SrTiO<sub>3</sub> system can possibly be used to understand the pairing mechanism of cuprates such as BSCCO because two systems have the similar structure consisting of a charge-reservoir layer

(STO in the present case and BiSrO in BSCCO) and a superconducting layer (FeSe in the present case and CuO<sub>2</sub> in BSCOO).

The embodiments shown and described above are only examples. Even though numerous characteristics and advantages of the present technology have been set forth in the foregoing description, together with details of the structure and function of the present disclosure, the disclosure is illustrative only, and changes may be made in the detail, including in matters of shape, size and arrangement of the parts within the principles of the present disclosure up to, and including, the full extent established by the broad general meaning of the terms used in the claims.

What is claimed is:

1. A method for making a high-temperature superconducting film, comprising:

(S1) loading a SrTiO<sub>3</sub> substrate into an ultra-high vacuum system;

(S2) growing a single crystalline FeSe layer on a surface of the SrTiO<sub>3</sub> substrate by molecular beam epitaxy; and

(S3) growing a protective layer with a layered crystal structure by molecular beam epitaxy and covering the single crystalline FeSe layer by the protective layer.

2. The method of claim 1, wherein the protective layer is a single crystalline FeTe layer.

3. The method of claim 2, wherein the SrTiO<sub>3</sub> substrate is held at a temperature in a range from about 310 degrees Celsius to about 330 degrees Celsius in the process of growing the single crystalline FeTe layer.

4. The method of claim 3, further comprising coevaporating a Fe source and a Te source in the step (S3), wherein an evaporated temperature of the Fe source is in a range from about 1000 degrees Celsius to about 1100 degrees Celsius, and an evaporated temperature of the Te source is in a range from about 240 degrees Celsius to about 280 degrees Celsius.

5. The method of claim 2, wherein a thickness of the single crystalline FeTe layer is in a range from about 2 unit-cells to about 10 unit-cells thick, and a thickness of the FeSe single crystalline layer is in a range from about 1 unit-cell to about 5 unit-cells thick.

6. The method of claim 1, further comprising etching the SrTiO<sub>3</sub> substrate by hydrochloric acid for about 30 minutes to about 60 minutes and annealing in oxygen atmosphere for about 2.5 hours to about 3.5 hours at a temperature from about 950 degrees Celsius to about 1000 degrees Celsius before loading the SrTiO<sub>3</sub> substrate into the ultra-high vacuum system in the step (S1).

7. The method of claim 1, further comprising degassing the SrTiO<sub>3</sub> substrate at a temperature from about 600 degrees Celsius to about 650 degrees Celsius and keeping the temperature for about 2.5 hours to about 3.5 hours before growing the single crystalline FeSe layer in the step (S2).

8. The method of claim 1, wherein in the step (S2), the SrTiO<sub>3</sub> substrate is held at a temperature of in a range from about 380 degrees Celsius to about 420 degrees Celsius in the process of growing the single crystalline FeSe layer.

9. The method of claim 1, further comprising coevaporating a Fe source and a Se source in the step (S2), wherein an evaporated temperature of the Fe source is in a range from about 1000 degrees Celsius to about 1100 degrees Celsius, and an evaporated temperature of the Se source is in a range from about 130 degrees Celsius to about 150 degrees Celsius.

10. The method of claim 1, wherein in the step (S2), after growing the single crystalline FeSe layer, an entirety comprising the SrTiO<sub>3</sub> substrate and the single crystalline FeSe

layer is annealed at a temperature from about 450 degrees Celsius to about 550 degrees Celsius for about 20 hours to about 30 hours.

11. A method for making a high-temperature superconducting film, comprising:

(S1) acid etching a SrTiO<sub>3</sub> substrate by hydrochloric acid for from about 30 minutes to about 60 minutes and annealing for from about 2.5 hours to about 3.5 hours at a temperature from about 950 degrees Celsius to about 1000 degrees Celsius;

(S2) loading the SrTiO<sub>3</sub> substrate in an ultra-high vacuum system;

(S3) growing a single crystalline FeSe layer on a surface of the SrTiO<sub>3</sub> substrate by molecular beam epitaxy; and

(S4) growing a protective layer with a layered crystal structure by molecular beam epitaxy and covering the single crystalline FeSe layer by the protective layer.

12. The method of claim 11, wherein the protective layer is a single crystalline FeTe layer.

13. The method of claim 12, wherein the SrTiO<sub>3</sub> substrate is held at a temperature in a range from about 310 degrees Celsius to about 330 degrees Celsius in the process of growing the single crystalline FeTe layer.

14. The method of claim 13, further comprising coevaporating a Fe source and a Te source in the step (S4), wherein an evaporated temperature of the Fe source is in a range from about 1000 degrees Celsius to about 1100 degrees Celsius, and an evaporated temperature of the Te source is in a range from about 240 degrees Celsius to about 280 degrees Celsius.

15. The method of claim 12, wherein a thickness of the single crystalline FeTe layer is in a range from about 2 unit-cells to about 10 unit-cells thick, and a thickness of the FeSe single crystalline layer is in a range from about 1 unit-cell to about 5 unit-cells thick.

16. The method of claim 11, further comprising degassing the SrTiO<sub>3</sub> substrate at a temperature from about 600 degrees Celsius to about 650 degrees Celsius and keeping the temperature for from about 2.5 hours to about 3.5 hours before growing the single crystalline FeSe layer in the step (S3).

17. The method of claim 11, wherein in the step (S3), the SrTiO<sub>3</sub> substrate is held at a temperature in a range from about 380 degrees Celsius to about 420 degrees Celsius in the process of growing the single crystalline FeSe layer.

18. The method of claim 11, further comprising coevaporating a Fe source and a Se source in the step (S3), wherein an evaporated temperature of the Fe source is in a range from about 1000 degrees Celsius to about 1100 degrees Celsius, and an evaporated temperature of the Se source is in a range from about 130 degrees Celsius to about 150 degrees Celsius.

19. The method of claim 11, wherein in the step (S3), after growing the single crystalline FeSe layer, an entirety comprising the SrTiO<sub>3</sub> substrate and the single crystalline FeSe layer is annealed at a temperature from about 450 degrees Celsius to about 550 degrees Celsius for about 20 hours to about 30 hours.

20. A method for making a high-temperature superconducting film, comprising:

(S1) loading a SrTiO<sub>3</sub> substrate in an ultra-high vacuum system;

(S2) growing a single crystalline FeSe layer on a surface of the SrTiO<sub>3</sub> substrate by molecular beam epitaxy; and

(S3) growing a single crystalline FeTe layer by molecular beam epitaxy and covering the single crystalline FeSe layer by the single crystalline FeTe layer.